Mercury contamination in remote forest and aquatic ecosystems in the Northeastern U.S.: sources, transformations and management options

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1.0 Introduction

1.1 Summary of the mercury issue

Mercury (Hg) is a potent neurotoxin of significant ecological and public health concern. Exposure to Hg occurs largely by consumption of contaminated fish. Indeed, children and women of childbearing age who consume large amounts of fish are at high risk (NAS 2000). It is estimated that over 630,000 children born each year are exposed to elevated methyl mercury (MeHg) in the womb and at risk for impaired neurological development (Mahaffey 2004). Eight percent of U.S. women of childbearing age have blood Hg levels in excess those deemed safe by the U.S. Environmental Protection Agency (USEPA) (Shober 2003). To protect the human population, the EPA established a MeHg criterion for fish tissue at 0.3 µg/g under Section 304(a) of the Clean Water Act (EPA 2001). As of 2004, fish consumption advisories due to Hg contamination have been issued for 45 states, including 19 statewide advisories. These advisories represent more than 100,000 lakes, 850,000 miles of rivers and 75% of the coastal waters, including all the Great Lakes (CTD check to see if these values are correct). Moreover, a healthy fishing resource is important to the economy. Fishing contributed $3 billion in economic output in the northeastern U.S. in 2001 (NESCAUM 2005). The extent of fish consumption advisories underscores the human and ecological health risk posed by Hg pollution.

1.2 Sources of mercury

Large Hg sources include electric utilities, incinerators, industrial manufacturing, wastewater treatment plants and consumer products (e.g. batteries, fluorescent light bulbs, Hg switches). There is considerable public policy...
attention directed toward airborne Hg emissions since they constitute the largest category of
global Hg sources (UNEP 2002). U.S. emissions of Hg peaked in the 1970s and have declined in
recent years (Pirrone et al. 1998). Nevertheless, Hg emissions to the atmosphere are
approximately three times higher than pre-anthropogenic levels (Mason et al. 1994).

1.3 Mercury and ecosystem health

Atmospheric emissions of Hg are not a direct public health risk. Mercury is transported
over variable distances (i.e., local to global scales), depending on the speciation of emissions and
reaction pathways in the atmosphere. Mercury is ultimately deposited to the landscape, largely in
an inorganic form. However, following deposition, inorganic Hg may be converted to a
bioavailable toxic form – MeHg -- which strongly bioaccumulates at all levels of the aquatic
food chain. This air-to-water-to-fish/biota linkage of Hg has challenged state and federal
regulators to control airborne emissions in order to limit Hg deposition to levels that will support
water and fish tissue standards. Achieving Hg reductions to decrease contamination of fish in
public waters requires a thorough understanding of the complex processes that link Hg through
air, watersheds, water, aquatic biota, wildlife and humans.

1.4 Objectives of this paper

To address this need, the Hubbard Brook Research Foundation (HBRF) has convened a
team of eight scientists to synthesize existing data from the northeastern U.S. and identify
sources, transformations, effects and management options for atmospheric Hg pollution. This
analysis was facilitated by a recent compilation and analysis of Hg data for eastern North
America as part of a Northeast Ecosystem Research Cooperative (NERC) initiative (Evers and
Clair 2005). As part of the HBRF effort, we also evaluate specific control scenarios for airborne
Hg emissions. In this synthesis we address a series of questions concerning: 1) Hg inputs, 2) Hg transport, transformations and effects, and 3) Hg policy and management in the northeastern U.S.

1.4.1 Policy context

Efforts are underway at state, regional, national and global levels to further reduce Hg emissions. In the 1990s the U.S. issued regulations to limit emissions from certain waste incinerators by more than 90%. The New England Governors and Eastern Canadian Premiers adopted an action plan to reduce Hg emissions by 75% within the region by 2007 (NEG-ECP). In June 2005 the USEPA adopted a rule pertaining to Hg emissions from coal-fired power plants. This rule calls for a two-phase reduction in emissions that is predicted to result in a 69% decrease in total U.S. emissions from electric utilities by 2020 using a cap and trade approach. Other national proposals called for a 70 to 95% reduction in emissions from power plants. Some proposals proposed Hg trading within a cap and trade program, while others required all plants to meet a specified emissions level. With the final passage of the USEPA rule, all 50 state are required to develop USEPA-approved state implantation plans to ensure that Hg emissions levels will be achieved. The debate over target reduction levels and cap and trade approaches will likely reoccur at the state level as these plans are developed.

1.4.2 Northeast study region

The northeastern U.S. is a compelling region to investigate inputs, transformations, effects and management options of Hg. The region is largely forested with a large number of intact wetlands and encompasses centers of high human population density along the coast. As such, it includes a range of ecosystems from forests in rural uplands, agricultural lands along river valleys, and urban and suburban watersheds draining to coastal estuaries. The region includes an airshed with elevated Hg emissions and ecosystems that are particularly sensitive to
Hg inputs, such as high elevation forested areas with shallow surficial materials, an abundance of wetlands and unproductive surface waters. A combination of high Hg deposition and sensitive ecosystems, results in lakes and streams with elevated Hg in fish and wildlife across the Northeast landscape. These biological “hotspots” require special attention from both a scientific and policy perspective. A “hotspot” is a term used to describe known areas with elevated Hg deposition, and/or Hg concentrations in fish and wildlife that exceed adverse effects criteria. We address causes and implications of Hg “hotspots” in a companion paper (Evers et al. this issue)

2.0 What are the sources of Hg emissions and pathways of Hg deposition to ecosystems of the northeastern U.S.?

2.1 Emissions of Hg

2.1.1 Historic emissions by source category for global, U.S., Northeast

Current estimates of global Hg emissions range from 6,060 to 6,600 metric tons/yr, with 2,143 to 2400 tons/yr (33-36%) attributed to direct anthropogenic emissions (NESCAUM 2003). The remaining 4,000 tons/yr are derived from natural sources or re-released from past anthropogenic emissions (Bergan 1999; Mason and Sheu 2004; Segneur et al. 2004; Lamborg et al. 2002). These values suggest that about two thirds of atmospheric emissions of Hg are derived from either direct or re-emitted anthropogenic sources. Considerable Hg is emitted from stationary combustion sources (1,450 tons/yr; 50% of anthropogenic sources; Pacyna et al. 2003). Total U.S. anthropogenic emissions from all sources are estimated at 120 ton/yr, with the Northeast contributing about 6.3 ton/yr (National Emission Inventory, 1999).

Mercury emissions have decreased markedly in the U.S. over the past decade (Table 1). These decreases have been primarily due to large reductions in medical waste and municipal incinerator emissions associated with federal regulations (EPA 2003). Conversely, emissions
from electric utilities have remained largely unchanged and their relative contribution to U.S. emissions has increased from 25% to 40% over the past decade. The next largest category in the U.S. is combined emissions from industrial, commercial, institutional boilers and process heaters (12 tons/yr). In the Northeastern states the largest sources in the 2002 inventory are municipal incinerators, followed by coal electric utilities (NESCAUM 2005).

2.1.2 Speciation of mercury emissions

Mercury is emitted to the atmosphere from point sources in three forms: elemental Hg (Hgo), gaseous ionic Hg (reactive gaseous Hg; RGM) and particulate Hg (PHg). Knowledge of Hg speciation is critical to understanding the fate of atmospheric Hg emissions.

The speciation of Hg sources varies widely (Table 2; EPA 1999). Emissions of Hg species from point sources in northeastern U.S. in 1999 were 2.55, 1.50, and 0.45 ton/yr for Hgo, RGM, and PHg, respectively (National Emissions Inventory 1999). Several studies provide the average emission profile from coal combustion in the U.S. as roughly 50% Hgo, 40% RGM and 10% PHg (Pacyna et al. 2003, EPRI 2000). However, emissions from coal combustion in the Northeastern states have a higher proportion of RGM (68%) and lower percentage of Hgo (30%) and PHg (2%) (NESCAUM 2005).

The specific Hg emission speciation profile for a given power plant depends on the type of coal and the air pollution control technology used (NESCAUM 2003, Rossler 2002); in general Western coal utilities are much less numerous than Eastern coal utilities, burn Western coal which contains lower amounts of total Hg, and emit a greater percentage of elemental Hg than Eastern coal utilities (Figure 1). These factors contribute to the lower Hg deposition levels in the West compared to the East.

2.1.3 Residence time in the atmosphere
Hg can be a local, regional and global pollutant, depending on its speciation, due to a range of residence times in the atmosphere. Consequently, it can deposit locally or be transported long distances (Dastoor and Larocque 2004). The potential for some forms of Hg to act as a global pollutant does not obviate the need for local, and regional control efforts to decrease Hg deposition (Engstrom and Swain 1997).

Elemental Hg is by far the largest pool of Hg in the atmosphere due to its relatively long residence time of 0.5 to 2 years. As a result, Hgo can be transported long distances (i.e., tens of thousands of kilometers). However, recent research shows that under some conditions Hgo can be rapidly converted to RGM and deposited locally or regionally, including in the Arctic and Antarctic atmospheres (Ebinghaus et al. 2002, Lindberg et al. 2002), in the marine and continental boundary layer and in urban plumes (Weiss-Penzias et al. 2003). Elemental Hg can also be directly deposited to forested ecosystems and be taken up through gas exchange within the stomata. These recent studies suggest that the atmospheric lifetime is Hg° is probably closer to 0.5 years than to 2 years.

Reactive gaseous Hg is predominantly gaseous chloride and oxide forms of ionic Hg. This species is highly soluble in water and readily deposited to surfaces within tens to a few hundreds of kilometers from emission sources, having an atmospheric residence time of 0.5-2 days, which results in enhanced deposition near sources.

The atmospheric residence time of PHg is also short (0.5-3 days). While the PHg fraction in ambient air in rural and remote areas is small relative to Hgo° (generally less than 5%; Schroeder and Jackson, 1987; Horvat, 1996), it can be more abundant near Hg sources, where it may approach 50% of the total atmospheric Hg (Schroeder et al., 1991; Lamborg et al., 1995; Keeler et al., 1995). A recent study in the Arctic has found that the concentrations of PHg can,
under certain atmospheric conditions, be much higher than gaseous Hg in ambient air (Lu et al.
1998; Lu et al. 2001). Very recently, data collected in the Florida Everglades indicated that dry
deposited PHg significantly contributed to the total Hg deposition and to the levels of Hg in
runoff (Gildemeister 2001).

2.2 Atmospheric deposition of mercury

Although some sites experience Hg contamination due to local industry, solid waste
disposal or discharge from wastewater treatment plants, the predominant input of Hg to most
watersheds is atmospheric deposition, primarily of RGM and PHg (Lindqvist et al. 1991, Mason
Fitzgerald et al. (1998) systematically rule out alternate hypotheses, such as natural weathering
as contributing to widespread contamination of Hg.

Atmospheric deposition of Hg occurs in both wet and dry forms. Wet deposition refers to
Hg in rain, snow, fog and other forms of precipitation. Dry deposition includes Hg in gaseous
and particulate forms.

2.2.1 Historical trends in mercury deposition based on sediment cores

Unfortunately no accurate estimates exist of Hg deposition prior to 1990. However,
researchers using global models (Hudson et al. 1995), reconstructions of mass balances (Mason
et al. 1994) and paleolimnological techniques (Engstrom and Swain 1995) generally find that
deposition of Hg has increased two to three-fold over the past two centuries following increases
in Hg emissions associated with industrialization and Hg use (Engstrom and Swain 1995), with
some locations exhibiting greater than 20-fold increases (Meili 2003, Nriagu and Becker 2003,
Pirrone et al. 1998). Several paleolimnological studies have been conducted in the Northeast, all
showing similar patterns (Lorey and Driscoll 1999; Kamman and Engstrom 2002; Perry et al
Deposition of Hg increased starting in the late 1800s to early 1900s, increasing 2.5 to 15 fold by the 1970s to 1990s. Modest decreases in sediment Hg deposition have been evident in recent years, but values remain well above pre-industrial values (2.5 to 5 fold).

2.2.2 Local vs. regional vs. global contributions to deposition

The U.S. receives considerable deposition of Hg (EPA 1997). Identifying the specific sources of this Hg is difficult. Nevertheless, most research indicates that Hg sources in the U.S. are the largest contributor to Hg deposition in the U.S. Of the estimated 87 tons/yr of Hg deposited in the U.S. in 1997 (Tom Do we have a more current value? I don’t know of one YoungJi do you?), 35 tons (40%) are likely to originate from global sources and 52 tons (60%) from U.S. sources (Rossler 2002, EPA 1997). Of the 52 tons deposited in the U.S. from U.S. sources, at least 24 tons (28%) are likely to originate from domestic utility coal boilers (48 tons of emitted Hg x 0.5 fraction likely to be deposited within the U.S.).

Similarly, for the Adirondacks, Catskills and Finger Lakes regions of New York it is estimated that 11 to 21% of the Hg deposited was derived from emissions within New York State, 25 to 49% originated from other U.S. sources, and 13 to 19% originated from Asia (NYSERDA).

2.2.3 Spatial and temporal patterns of wet deposition in U.S., Northeast, local areas

In the U.S. and Canada, measurements of wet Hg deposition are largely made through the Mercury Deposition Network (MDN). Across North America, wet Hg deposition is highest in the Southeastern U.S. (i.e., Florida, Mississippi). There are currently six MDN sites in the Northeast (NY, NH, ME), with annual deposition ranging from 3.8 to 12.6 µg/m²-yr (Figure 3: VanArsdale et al. 2005). The network is sparse but there does not appear to be broad spatial patterns in wet Hg deposition across the region. A seasonal pattern for Hg in precipitation in an
11-year event deposition record at Underhill VT is clearly evident, with higher Hg concentrations and deposition observed during spring and summer months due to a mix of meteorological, source emissions, and atmospheric chemistry influences.

2.2.4 Estimates of dry deposition in the Northeast

Estimates of dry deposition are highly uncertain because of the complex dependence on atmospheric conditions, collection surface characteristics, and chemical properties. Several regional and local modeling efforts have been conducted to estimate the amount of dry deposition of Hg in U.S. Recently, a 3-D Eulerian model was applied to North America to simulate dry deposition of RGM and PHg. In the Adirondacks, Finger Lakes, and Catskills regions of New York estimated dry Hg deposition was 7.0, 3.9, and 10.2 \( \mu g/m^2\)-yr, respectively (Seigneur et al. 2003). Another model estimate specifically for Northeast suggests that dry deposition of RGM plus Hg\(^0\) was 37.1 \( \mu g/m^2\)-yr (Xu et al. 2000). Both studies indicate that dry deposition is a significant pathway of Hg inputs (50 to 75% of total deposition), and agree with EPA predictions that regionally Hg dry deposition in Northeast is the highest in the country. These high inputs are in part related to the abundant forests that are particularly effective at collecting dry deposition via the canopy.

2.2.5 Deposition to a local area

Given that most combustion sources emit some fraction of Hg as RGM and PHg, it is anticipated that a portion of the emissions will deposit in the area near these sources (local deposition). An analysis of emissions and deposition in southern New Hampshire shows a local region of high deposition in the vicinity of Concord, NH. Most of the Hg deposited in that area originated from sources within a 50 km radius (Evers et al, this issue). Moreover, a review of Hg concentrations in yellow perch (\textit{Perca flavescens}) and common loons (\textit{Gavia immer}) in this
same area indicates that a biological hotspot of Hg exists in the same area (Evers et al, this issue). It appears that the amount of Hg deposited to this area and found in common loons declined as surrounding point sources reduced Hg emissions approximately 58% between 1996 and 2002 (Han et al. in prep; Evers et al, this issue).

2.2.6 Canopy enhancement – throughfall and litterfall

Due to the large surface area associated with canopy foliage, atmospheric deposition of contaminants is elevated in forests compared to other types of ecosystems. Several forest studies have indicated that total atmospheric Hg deposition may be estimated from fluxes of throughfall plus litterfall (Driscoll et al. 1994; Johnson and Lindberg 1995; Lindberg 1996; Rea et al. 2001). Throughfall is precipitation that passes through the canopy leaching accumulated dry deposition. Most studies show that throughfall fluxes of Hg greatly exceed wet deposition (Grigal 2002). Litterfall is deposition of leaf, needle and other plant materials to the forest floor.

At most sites the major flux of Hg deposited to the forest floor is litterfall, which greatly exceeds wet deposition (Driscoll et al. 1994; Lindberg 1996; Rea et al. 2001). In a review of the literature Grigal (2002) suggests that the ratio of wet deposition to throughfall to litterfall Hg fluxes is 1:1.8:2.2. So for 5 µg/m²-yr of wet deposition that might be typical of the Northeast, anticipated throughfall would be 9 µg/m²-yr and litterfall would be 11 µg/m²-yr for total Hg deposition of 25 µg/m²-yr and dry deposition of 19 µg/m²-yr (75% of total deposition).

Within the Northeast a recent modeling effort found that spatial patterns of total Hg deposition at a regional scale are complex (Miller et al. 2005). In general, total Hg deposition was highest in the southwest and lower in the northeast portion of the region, although elevation, land cover and proximity to urban areas modified this general pattern. The estimated net Hg° and RGM fluxes enhanced by forest vegetation were each generally greater than wet deposition,
suggesting that vapor phase Hg assimilation by plant foliage and subsequent litterfall may be a substantial Hg input to forest ecosystems (Miller et al. 2005).

2.2.7 Re-emission

Some portion of Hg deposited into the surface is re-emitted into the atmosphere. Mercury volatilization from soil involves three steps: 1) biotic or abiotic reduction of ionic Hg to Hg⁰, 2) diffusion of Hg⁰ to the soil surface and 3) diffusion of Hg⁰ across the soil-air boundary layer to the atmosphere (Zhang and Lindberg 1999). There have been a few studies of volatilization of Hg from soil, showing a wide range of rates (Grigal 2002). In general, volatilization rates are high immediately following inputs of ionic Hg to soil (Schluter et al. 1995; Grigal 2002). Based on a review of the literature, Grigal (2002) estimated a mean rate of Hg⁰ volatilization from soil over a growing season of 32µg/m²-yr. This rate is more than adequate to re-emit atmospheric Hg deposition. The magnitude and uncertainty of this process demonstrates the acute need for additional research on soil Hg emissions.

3.0 How is Hg transported in forest-wetland-lake ecosystems and what are the transformations and fate of these Hg inputs?

Following Hg deposition to the landscape, it may be sequestered in soil, re-emitted back to the atmosphere or transported through the watershed, with a fraction ultimately supplied to surface waters. Freshwater aquatic ecosystems are among the most sensitive to Hg pollution and Hg concentrations in surface waters of the Northeast vary by an order of magnitude (<0.51 to 12.7 ng/L; 5-95 percentile; Dennis et al. 2005; Figure 4). Ecosystem characteristics are also important in determining both the production and bioaccumulation of MeHg in surface waters. Under conditions of high total Hg loading, a range of MeHg bioaccumulation can occur
depending on the efficiency of the methylation and bioconcentration processes (Krabbenhoft et al. 1999).

3.1 Mercury transport and fate in upland forest ecosystems

Immobilization of Hg in forest soil, or the extent to which it remains sequestered in soil, appears to coincide with deposition of organic carbon (Schuster 1991; Schwesig et al. 1999). Concentrations of Hg in soil are generally strongly correlated with concentrations of organic carbon. There have been few direct studies of soil sequestration of Hg. Pools of Hg in upland soil in northern temperate regions are about 8 mg/m² (Lindqvist et al. 1991; Nater and Grigal 1992), although higher levels have been reported in central Europe (Schwesig and Matzner 2000).

The export of Hg from waters draining upland soils is generally low. As in soil, concentrations and fluxes of Hg in soil waters are closely related to dissolved organic carbon (DOC; Dennis et al. 2005). In northern forests, concentrations of total Hg are highest in waters draining the upper soil, coinciding with high concentrations of DOC. Concentrations and fluxes of total Hg decrease as DOC is immobilized with depth in mineral soil (Grigal 2002).

There are few studies of MeHg in upland soils or soil waters. Concentrations in soil and ground waters are generally low (0.02-0.2 ng/L), although studies generally show higher concentrations in upper soil waters and decreases with soil depth (Grigal 2002). There are few measurements of methylation rates in upland soils. However, low concentrations and fluxes of MeHg in drainage waters suggest that rates of methylation are generally low and freely draining upland soils are not important in the supply of MeHg to downstream surface waters, with the possible exception of recently harvested upland forests (Porvari et al. 2003).
3.2 Transport and transformation of Hg in wetlands

Wetlands are important features of the landscape that control the supply and speciation of Hg to adjacent surface waters. Although small amounts of MeHg enter terrestrial ecosystems by atmospheric deposition (Fitzgerald et al. 1991; St. Louis et al. 1994), concentrations of MeHg greatly increase in waters draining wetlands (Krabbenhoft et al. 1995; Branfireun et al. 1996). Annual rates of MeHg production in wetlands are approximately 0.3-0.5 $\mu$g/m²-yr (Rudd 1995). The factors controlling methylation of Hg in wetlands are not clear, but likely involve amounts and types of organic acids, hydrologic flow paths, as well as rates of microbial activity (Watras et al. 1995). Wetlands are a major source of DOC (Eckhardt and Moore 1990). Organic acids produced in wetlands complex both ionic Hg and MeHg enhancing the transport but decreasing the bioavailability of these species (Hudson et al. 1994). Further, discharge of water through wetlands, in part, determines the distribution and abundance of sulfate-reducing bacteria, which appear to be largely responsible for Hg methylation (Gilmour and Henry 1991; Choi et al. 1994a,b). Concentrations of MeHg from wetlands vary seasonally, with the highest concentrations evident during the summer due to warmer temperatures (and presumably greater rates of microbial activity) and longer hydraulic residence times (Bishop et al. 1995; Branfireum et al. 1996).

3.2.1 Transformations in surface waters

Mercury enters remote surface waters by direct atmospheric deposition, or by soil water, wetland or groundwater drainage. Stream and rivers often exhibit marked temporal variation in Hg concentrations often associated with variations in concentrations of DOC or suspended matter. Large increases in Hg concentrations can occur associated with hydrologic events (Shanley et al. 2005). Lakes are typically net sinks of total Hg, as losses from the water column
occur by sediment deposition and volatilization. In freshwater lakes, photochemical processes are largely responsible for the reduction of ionic Hg to Hg⁰ (Amyot et al. 1997). Microbial reduction has been shown in laboratory studies but at higher than natural concentrations of Hg (Morel et al. 1998). Lakes also result in net production of MeHg due to methylation in sediments and the water column.

Landscape patterns in average lake and river Hg concentrations are shown in Figure 4 for 18-minute grid cells as natural breaks in the range of average cell concentrations. The geographic distribution of average concentrations shows both the landscape-level heterogeneity in water Hg concentrations and several areas where concentrations are elevated across several contiguous cells (e.g., Adirondacks, southern New Hampshire, southern Nova Scotia). These areas are generally associated with high concentrations of DOC; rivers along Lake Champlain due to high suspended solids; and lakes in southern New Hampshire and eastern Massachusetts, presumably due to elevated atmospheric deposition.

Within the areas with the greatest mean Hg concentrations, within-cell heterogeneity is also greatest (Figure 4, legend E and inset), suggesting that even in areas with high Hg in surface waters, significant variation exists. This variation could result from high-flow events in rivers, which is evident in northeast Vermont and in the Champlain Valley (Gao et al., 2005; Shanley et al., 2005).

3.3 Other factors controlling Hg dynamics in surface waters

Other landscape attributes and watershed disturbances alter the transport, transformations and bioavailability of Hg.
3.3.1 Acidity, acid neutralizing capacity and sulfate

There are important linkages between Hg dynamics and acidic deposition. The Northeast receives elevated loading of acidic deposition as well as Hg, and contains a relatively large number of acid-impacted surface waters (Driscoll et al. 2001). Acidic deposition and the associated sulfur alter the acid-base status of surface waters and influence the transformations of Hg and Hg accumulation in fish.

Sulfur transformations are closed coupled with Hg dynamics. The solubility of Hg increases with increasing sulfide concentrations in anoxic waters though complexation reactions. Sulfide can also immobilize Hg by chemical precipitation (ref). Moreover, given that sulfate-reducing bacteria largely mediate Hg methylation, increases in sulfate up to a particular level increase concentrations of MeHg (Gilmour and Henry 1991; Gilmour et al. 1992). At high sulfate concentrations, MeHg concentrations decrease, presumably due to precipitation with sulfide. Experimental addition of sulfate to wetlands results in increases in methylation and export of MeHg (Branfireun et al. 1999, 2001; Swain et al. 2003). This pattern might infer that reductions in sulfur dioxide emissions and consequently sulfate deposition would result in decreased MeHg in the fish of receiving waters.

There have been widespread observations of increases in fish Hg concentrations with decreases in surface water pH (Grieb et al. 1990; Kamman et al. 2004). Hrabik and Watras (2002) used reference data and data from an experimentally acidified lake to examine the relative contribution of atmospheric Hg deposition and acidic deposition in regulating decreases in fish Hg concentrations. They found that one-half of the decrease in fish Hg over a six-year period could be attributed to decreases in acid loading (Hrabik and Watras 2002). This study suggests
lake acidification from acidic deposition has resulted in higher fish Hg concentrations than what would be expected under natural conditions.

3.3.2 Land use and watershed disturbance

In a study of 21 basins nationwide, watersheds with mixed agriculture and forest land cover had the highest methylation efficiency (Krabbenhoft et al. 1999). Even where these watersheds had low total Hg in sediments, they showed the highest MeHg in both sediment and water. Waters draining largely agricultural lands have been shown to have relatively high Hg concentrations of total Hg and MeHg, but lower concentrations in fish, presumably due to “bloom dilution” associated with high phosphorus loading (Kamman et al. 2004; see 3.4.2.1). Forest cutting has been shown to increase export of total Hg and MeHg (Porvari et al. 2003). Fire results in a complex pattern of Hg loss. During and shortly after fire elevated Hg losses appear to be associated with volatilization and drainage losses (Grigal 2002). Over the longer-term Hg transport to surface waters is reduced due to reductions in soil carbon and DOC concentrations.

Rates of Hg methylation can be altered by the fluctuation of reservoir levels associated with hydropower production. Many large waters in the Northeast are impounded to provide increased storage and/or daily peaking capacity. As a result, some reservoirs may fluctuate many centimeters on a daily basis or several meters over the course of a year. As the littoral zone experiences periodic wetting and drying associated with lake level fluctuation, varying cycles of reduction and oxidation enhance the production of MeHg (Weiner et al. 2003).

3.4 Bioaccumulation and trophic transfer of mercury

The relationship between total Hg or MeHg concentrations in surface waters and the Hg content of aquatic biota, such as fish, is non-linear. Factors controlling the bioaccumulation of
Hg in individual organisms and bioconcentration of Hg up the food chain are key to determining the likelihood of high MeHg in fish and other aquatic organisms.

3.4.1 Bioconcentration processes

Bioaccumulation and trophic transfer of Hg in aquatic food webs begins with bioconcentration of ionic and MeHg by primary producers. Bioaccumulation factors from water to algae at this trophic step are by far higher (≈10⁵-10⁶) than subsequent trophic levels (Plourde et al. 1997; Watras et al. 1998; Miles et al. 2001; Figure 5). Although both ionic and MeHg are taken up and diffuse through algal cell membranes, assimilation of MeHg is four times more efficient than ionic Hg (Mason et al. 1996). Aquatic grazers and predators take up MeHg mainly from their food rather than from water (Harris et al. 1993; Rodgers 1994; Hall et al. 1997; Harris and Bodaly 1998). Methyl Hg is efficiently transferred to the higher levels of the food web and retained in the fatty tissue of higher-level organisms. However, storage of MeHg in fish is largely in the muscle tissue of the filets, which distinguishes Hg from other biomagnified contaminants (e.g. PCBs, DDT) and makes human consumption more likely.

Methyl Hg in aquatic consumers and predators increases in concentration and comprises a greater percentage of the total Hg concentration as it progresses up the food chain (Figure 5). Methyl Hg comprises on average less than 10% of the total in water, averages 15% in phytoplankton, 9-95% in aquatic invertebrates (zooplankton, insects), and comprises most of the total Hg (>95%) in fish (Back and Watras 1995; Tremblay et al. 1996; Watras et al. 1998; Simon and Boudou 2001). Thus organisms consuming higher trophic level prey are exposed to higher concentrations of total Hg and MeHg (Cabana and Rasmussen 1994; Kidd et al. 1996; Vander Zanden and Rasmussen 1996).

3.4.2 Controls on the bioconcentration of mercury in aquatic food chains
A variety of physical, chemical and biological factors influence the bioaccumulation of Hg. Fish Hg concentrations tend to vary with lake or watershed area (positively correlated) and land use (Chen et al. 2005). These significant correlations are indicative of the biological and chemical complexity of processes that influence the availability of Hg species, Hg uptake by aquatic microbes and algae, and Hg transfer to higher trophic levels.

3.4.2.1 Nutrients and zooplankton abundance

The methylation of Hg in sediments of high nutrient ecosystems appears to be lower than in unenriched ecosystems resulting in lower MeHg concentrations in biota (Why? T.H.). Nutrient enrichment also diminishes Hg bioaccumulation in phytoplankton via the biodilution of Hg under algal bloom conditions (Pickhardt et al. 2002; Chen and Folt in press). Mercury concentrations in zooplankton also decrease with increasing zooplankton densities that in turn, are correlated with lower Hg concentrations in fish (Chen and Folt in press).

3.4.2.2 Trophic level and food chain length

Within given fish populations, Hg burdens increase with age and size of individuals in part due to the slower rates of elimination in larger individuals and the consumption of higher trophic level foods by older and larger individuals (Wiener and Spry 1996; Trudel and Rasmussen 1997). Mercury concentrations in top predator fish are higher in food webs with longer chain lengths and fewer linkages (Cabana and Rasmussen 1994; Cabana et al. 1994; Stemberger and Chen 1998). This mechanism likely explains why fish feeding from longer pelagic food webs carry higher Hg burdens than benthic fish feeding on shorter food chains (Becker and Bigham 19XX). This last sentence seems to be repetitive (T.H.)
3.4.2.3 Bioavailability of “new” versus “old” mercury

Experiments conducted in two very different watersheds in Ontario and the Florida Everglades suggest that Hg that is newly added through atmospheric emissions and deposition is more likely to accumulate in the food chain than previously deposited Hg (Atkeson et al. 2003, Blanchfield unpublished data, Gilmour et al. 2001, Gilmour et al. 2003).

An experimental study (METAALICUS) is being conducted at the Experimental Lakes Area in Ontario, Canada in which stable Hg isotopes are being added to a whole ecosystem (lake and watershed) to examine the response in fish Hg concentrations. The first two years of results show that Hg added directly to a lake surface is readily converted to MeHg and can be observed in fish in the same season that the additions are made. After 2 years, approximately one quarter of the Hg in young perch was due to Hg added to the lake surface (Blanchfield, unpublished data). Furthermore, the Hg added to the lake surface each year appears to be more available for conversion to MeHg (and then move through 2003; Celia I’m not sure what you mean by this).

Hrabik and Watras (2002) reported a rapid decline in fish Hg as Hg inputs to Wisconsin lakes declined. Based on this and other information, they concluded that even with a large reservoir of Hg in lake sediments and in decomposing organisms, newly deposited Hg dominated bioaccumulation processes in their study lakes.

3.5 Indicators of mercury sensitivity

Often fish Hg data are not widely available to resource managers to evaluate and predict the extent and likelihood of Hg contamination for a state or region. As discussed above, Hg bioaccumulation in fish tissues is related to several lake chemical attributes, including DOC, pH, acid neutralizing capacity (ANC) and total phosphorus. We combined data from three stratified,
random-probability surveys of northeastern lakes (USEPA EMAP Northeast Lakes Program, 1991-1994; Maine REMAP program 1994-1995; VT-NH REMAP Program, 1998-2000), with the survey datasets of Chen et al. (2005) to examine the characteristics of water chemistry in lakes with high (> 0.3 µg/g) and low (<0.3 µg/g) mean concentrations of Hg of standard length yellow perch (Figure 6). This analysis showed that lakes containing yellow perch with high Hg concentrations (>0.3 µg/g) had significantly higher DOC, and lower pH, ANC and total phosphorus concentrations than lakes with low (<0.3 µg/g) fish Hg concentrations.

Using this statistical analysis, we established Hg sensitivity indices for northeastern U.S. lakes (Table 3). Approximately 75% the survey lakes in the region with DOC > 4.0 mg C/L had pH <6.0 and ANC < 100 µeq/L had yellow perch Hg > 0.3 µg/g. The threshold value for total phosphorus was 30 µg P/L, and 58% of lakes across the region were above this value. This analysis suggests that a high percentage of lakes in the Northeast have chemical characteristics conducive to high fish Hg concentrations.

3.6 Mercury exposure and effects on biota

3.6.1 Spatial patterns

Long-term sampling of Hg in common loons across North America show a west to east spatial pattern of MeHg availability, with northeastern North America exhibiting the highest levels (Evers et al. 1998, 2003; Scheuhammer et al. 2001). Within the Northeast, extensive fish, bird and mammal sampling efforts documented areas with Hg concentrations in biota exceeded generally accepted effects thresholds (Evers 2005; Evers et al. this issue). Most of these “biological hotspots” were distant from point sources and received Hg largely via atmospheric deposition. In addition, point source discharges to surface waters are common and include areas such as on the Sudbury River in Massachusetts and the Penobscot River in Maine.
3.6.2 Phylogenetic patterns

Biota are exposed to MeHg primarily through the food they consume. Recent efforts in the Northeast have established robust Hg exposure profiles for multiple species of fish, birds, and mammals (Table 4; Evers and Clair 2005). These spatially-associated (spatially-explicit?) data provide insight on the phylogenetic relationships of MeHg availability and substantiate the importance of incorporating important confounding factors including habitat type, foraging guilds, trophic structure, and demographic variables (Evers et al. 2005).

3.6.2.1 Aquatic invertebrates

Mercury burdens in aquatic benthic invertebrates and larval insects in the Northeast have been studied most extensively in lakes and reservoirs in Quebec (Tremblay et al. 1996, 1997, 1998). In general, Hg concentrations vary by taxonomic group of benthic organism with higher trophic levels containing higher proportion of MeHg (odonates > heropterans and coleopterans > trichopternas > dipterans and ephemeropterons (Tremblay et al. 1996). Comparisons revealed 3-10 times higher total Hg levels in larval insects from reservoirs than natural lakes (Tremblay et al. 1996). In crayfish, Pennuto et al. (2005) found the highest levels in headwater streams and in one study site were over five times higher than natural lakes.

3.6.2.2 Fish

Total and MeHg in fish have been measured across a variety of species, tissues, ages, and sizes of individuals. Fair comparisons among studies are therefore difficult to construct, although efforts to standardize species and normalize size and species differences are progressing. Fish provide an important biotic hinge in assessing ecosystem impacts or human health concerns. Kamman et al. (2005) summarized over 15,000
fish Hg levels from 24 separate surveys in the Northeast and found the yellow perch and brook trout (Salvelinus fontinalis) as species regularly sampled. These two species stand as good indicators for relating the risk of Hg to higher trophic level organisms, such as the common loon. Mean fillet Hg levels from 13 species in this dataset show highest levels in large fish and top predators (Figure 7). (Dave – can we make a general statement like the one above and then delete the result of this detail since it will be in the figure?)in walleye (Stizostedion vitreum), white perch (Morone americana), and lake trout (Salvelinus namaycush)and lowest levels in brown bullhead (Ictalurus nebulosus), white sucker (Catostomus commersonii), and brown trout (Salmo trutta) (Figure 7). The large fish Hg dataset that includes both whole body and fillet tissue Hg levels from 40 different species, now provides an effective way to assess the exposure and effects of Hg in fish and associated wildlife.

3.6.2.3 Reptiles and Amphibians

Mercury in herpetofauna is less studied than most other vertebrate taxons. Efforts with salamanders (Bank et al. 2005), frogs (Ugarte et al. 2005), snakes (Rainwater et al. 2005), and turtles (Golet and Haines 2001) indicate elevated uptake of MeHg. Bank et al. (2005) showed the importance of two-lined salamanders (Eurycea bistlineata) to indicate MeHg availability in headwater streams, while Bank et al. (2006) demonstrated with these salamanders and other amphibians how Hg levels can in headwater streams can be significantly different than those in receiving lakes. (Is this generally true or linked to a specific location i.e. Northeast? T.H.)

3.6.2.4 Birds

Bird Hg exposure levels have been studied in a wide range of species and efforts are underway to choose species that are suitable indicators of MeHg availability across North
Species from several foraging guilds are likely prone to MeHg availability that is more elevated than historically in the environment (confusing sentence). Piscivores have been the most thoroughly studied and include species likely at greatest risk including the common loon (Evers 2006), wading birds (Frederick et al. 1999), and the bald eagle (Bowerman et al. 2002). Other foraging guilds, such as those depending on invertebrates, have been recently demonstrated to also biomagnify MeHg to elevated levels. Evers et al. (2005) showed that some songbirds such as the northern waterthrush (Seiurus noveboracensis) and red-winged blackbird (Agelaius phonencius) can have blood Hg levels greater than associated piscivorous belted kingfishers (Ceryle alcyon). New findings by Rimmer et al. (2005) indicate that terrestrial insectivores, such as the Bicknell’s thrush (Catharus bicknelli) and other montane songbirds, are accumulating MeHg. Concurrent sampling indicate that levels are three times higher in that thrushes Caribbean wintering areas (something missing in last sentence).

3.6.2.5 Mammals

The most commonly studied terrestrial mammals for Hg exposure are mink and otter. Yates et al. (2005) established a Hg exposure profile for mink (Mustela vison) and river otter (Lotra canadensis) for northeastern North America. Mercury levels tended to be greater in mink than otter, interior versus coastal populations, and females versus males. Raccoons (Procyon lotor) have also been used as a relevant indicator species (Porcella et al. 2004) but their Hg body burdens can be confounded by their ominivorous diet and link with human habitations. The importance of different foraging guilds identified in birds extends to mammals. Piscivorous freshwater mammal species are traditionally the group of mammals most emphasized, however, recent evidence with MeHg biomagnification in insectivores has increased investigations into exposure in bats.
3.7 Mercury effects in biota (similar heading to 3.6)

Mercury assessments for birds generally target physiological, behavioral, and reproductive impacts in individuals. Often, studies are based on laboratory experiments for fish (Hammerschmidt et al. 2003, Webber and Haines 2003), birds (Heinz 1974, 1979; Spalding et al. 2000), and mammals (Aulerich et al 1974). Laboratory studies are useful tools for gaining insight on effects in the wild. In birds, lethargy has been linked to elevated MeHg levels in the laboratory and follow-up studies in the wild has shown that the common loon exhibits such behaviors when associated with elevated Hg exposure (Nocera and Taylor 1998), particularly when adults have blood Hg levels > 3.0 ug/g (ww) (Evers et al. 2004) or egg Hg levels > 1.3 ug/g (ww) (Evers et al. 2003). Multiple Hg exposure studies on the common loon across wide geographic areas and a recently developed population model (Evers 2006 – Dave, is this in prep. Or in press?) provides a robust platform for understanding the spatial and temporal trends of MeHg availability. Multiple studies on the effects of Hg and associated confounding factors (Merril et al. 2005) also provide a good backdrop for determining thresholds for the loon at a population level. Nacci et al. (2005) quantitatively demonstrate how an ecological risk assessment model using the common loon can be used for managing and identifying impacts of Hg on wildlife populations in the U.S.

For these reasons, the common loon is selected as our biotic indicator to evaluate the relationship between changes in Hg deposition associated with changes in MeHg availability based on our application of the Mercury Cycling Model for Headwater drainage lakes (MCM-HD; see section 4).
4.0 What are the management options for reducing mercury inputs?

At present most policy attention is focused on Hg emissions from power plants since they are the largest single source of atmospheric Hg in the U.S. Many proposals have been introduced at both the federal and state level to control Hg emissions from power plants. The main differences among them are: the level and timing of the cuts, the existence or absence of an emissions cap, and whether or not trading is allowed. In general, the level and timing of Hg emission reductions is likely to control the timing and extent of recovery, while the use of trading raises the question of the persistence or expansion of Hg “hotspots”.

We have used the MCM-HD to evaluate the relationship between reductions in Hg deposition and concentrations of Hg in surface water and yellow perch. The resulting output for yellow perch Hg is then used in a population model to estimate changes in population level risk to common loons.

The MCM-HD has been calibrated and applied to the Sunday Lake watershed in the Adirondack region of New York. (Munson et al.). Sunday Lake is a drainage lake, with extensive wetlands, an average pH of 5.5, ANC 20 μeq/L and DOC 10 mg C/L. Current Hg concentrations in yellow perch average 0.88 μg/g for 3+ to 5+ (w.w. fillet). The Sunday Lake watershed has a relatively large wetland area (20%) and a relatively large watershed to lake area ratio (~175). As a result of these characteristics Sunday Lake is very sensitive to atmospheric Hg deposition and probably not highly responsive to decreases in deposition.

The two scenarios evaluated for Sunday Lake were step reductions of total atmospheric Hg deposition by 15% and 50%. The 15% reduction in deposition corresponds to the effect of aggressive electric utility controls, whereas the 50% reduction scenario represents the potential effect of decreasing emissions from a wide range of sources. The results for these two scenarios
were compared to base case conditions, which assume emissions and deposition will continue at the current level (Table 5).

The results show that simulated Hg concentrations in water and fish decline relatively rapidly over the first 35 years of the simulation and then decline slowly over the remainder of the simulation period. The rate of decline is highly uncertain. After 50 years, the water and fish Hg concentrations remain high under both scenarios, with fish concentrations at 0.83 µg/g in 2050 with a 50% decline in Hg deposition beginning 2005. The fact that fish Hg concentrations remain well above the USEPA criterion under all scenarios reflect that fact the Sunday Lake has watershed and surface water chemistry conditions that make it extremely effective at producing MeHg.

An important uncertainty in the MCM-HD model is the response of Hg concentrations in runoff from terrestrial ecosystems in response to changes in atmospheric Hg deposition. Predictions of fish response times for ecosystems receiving most of their Hg from terrestrial runoff strongly depend on assumptions made regarding the watershed response and the bioavailability for methylation of this Hg source. Understanding the fate of soil Hg pools is critical to assessments of the response of ecosystems to changes in atmospheric Hg deposition. If Hg deposition is permanently sequestered in soil, it is anticipated that surface waters will respond relatively rapidly to decreases in atmospheric Hg deposition. Conversely, if soil Hg is desorbed from soil in response to decreases in atmospheric Hg deposition the response time of watersheds to controls of Hg emissions will be greatly delayed. If newly added Hg is more available for methylation, the response times would be expected to shorten compared to current simulations. In addition, one of the important controls on the amount of Hg that is transported to surface waters and converted to toxic MeHg is watershed retention. Data from Lorey and
Driscoll (1999) and Kamman and Engstrom (2002) indicate that retention of atmospheric Hg deposition in lake-watersheds in Vermont, New Hampshire and the Adirondacks of New York has declined over recent decades. While the specific mechanisms responsible for this trend have not yet been determined, the data suggest that these watersheds have become more sensitive to Hg deposition. In other words, each year a greater percentage of the Hg is transported to surface waters where it may contribute to adverse environmental effects.

The yellow perch results from the MCM-HD model were then used in a population model for the common loon (Evers et al. 2004). The results show that even with substantial reductions in Hg deposition, common loons on Sunday Lake would experience Hg concentrations in female blood of approximately 11.0 (units?). At these levels, population levels of common loon in Sunday Lake would likely decrease and would be expected to produce 17% fewer fledged young over their lifetime, compared with common loons with blood Hg concentrations below. (Dave OK? Please edit)

While the MCM-HD model should be applied and tested at other sites, these results for Sunday Lake suggest that deep cuts in Hg emissions and deposition would have important benefits to water and fish – even in aquatic ecosystems remote from Hg sources, that there is a connection between Hg deposition and fish response – even in extremely vulnerable watersheds, and that recovery to Hg levels that no longer pose a human health risk or population level risk to loons will be a long-term process in some ecosystems that will require substantial reductions in Hg emissions from multiple sources.

5.0 Conclusions

From this synthesis of Hg in the environment of the northeastern U.S. we can conclude that:
• Direct emissions of anthropogenic Hg to the atmosphere and re-emissions of previously deposited anthropogenic Hg comprise much of the Hg inputs to the atmosphere.

• Atmospheric Hg can be a local, regional and global pollutant depending on the form in which it exists. Although Hg\textsuperscript{0} is generally thought to be a global pollutant emerging research suggests that all species have the potential to contribute to Hg deposition at a local and/or regional scale

• The dry deposition of Hg is poorly understood although it appears that a substantial fraction of atmospheric Hg deposition occurs as dry deposition.

• Atmospheric deposition of Hg is elevated in the northeastern U.S. compared to preindustrial values and deposition has decreased over the last 20-30 years in response to decreases in Hg use and Hg emissions.

• Mercury that is deposited on the landscape is retained in soil, re-emitted to the atmosphere or exported with drainage waters. The extent to which these processes occur and conditions which affect them are poorly known.

• Wetlands facilitate the transport of Hg to downstream surface waters through the supply of DOC and promote the production of MeHg due to reducing conditions.

• Methyl Hg readily bio-concentrates up the aquatic food chain to a factor 10\textsuperscript{6} to 10\textsuperscript{7} in piscivorous fish and wildlife.

• A variety of factors influence the sensitivity of surface waters to elevated concentrations of Hg in fish, including high concentrations of dissolved organic carbon, acidic conditions (i.e., low pH, ANC), and low total phosphorus concentrations.
• Differences in the bioavailability of new versus existing Hg suggest that the timing of Hg emissions reductions is important. Since new Hg appears to be more bioavailable than pre-existing Hg, rapid reductions will help facilitate recovery.

• Mercury is elevated in piscivorous animals and some terrestrial birds in the Northeast, at concentrations that could have deleterious effects.

• Utility emission reductions make an important contribution, but in sensitive ecosystems such as Sunday Lake, they are not sufficient to achieve recovery to safe levels for fish consumption and loon protection within 50 years.
Table 1. National Hg emissions in the U.S. by source category from 1990 to 1996 (tons/yr) and Hg emissions in the Northeast region in 2002 (kg/yr)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Utility coal boilers</td>
<td>51</td>
<td>51</td>
<td>48</td>
<td>814.5</td>
</tr>
<tr>
<td>Medical waste incinerators</td>
<td>50</td>
<td>40</td>
<td>3</td>
<td>16.1</td>
</tr>
<tr>
<td>Municipal waste combustors</td>
<td>42</td>
<td>32</td>
<td>5</td>
<td>1098.7</td>
</tr>
<tr>
<td>Industrial/commercial/institutional boilers and process heaters</td>
<td>12</td>
<td>12</td>
<td>12</td>
<td>270.1</td>
</tr>
<tr>
<td>Chlorine production</td>
<td>10</td>
<td>8</td>
<td>7</td>
<td></td>
</tr>
<tr>
<td>Hazardous waste incineration</td>
<td>7</td>
<td>4</td>
<td>7</td>
<td>1.4</td>
</tr>
<tr>
<td>Portland cement manufacturing</td>
<td>4</td>
<td>6</td>
<td>2</td>
<td>50.8</td>
</tr>
<tr>
<td>Pulp &amp; paper production</td>
<td>2</td>
<td>4</td>
<td>2</td>
<td>4.7</td>
</tr>
<tr>
<td>Total (all categories)</td>
<td>195</td>
<td>185</td>
<td>120</td>
<td>3573.2</td>
</tr>
</tbody>
</table>


Table 2. Mercury speciation ratio by source category (NEI, 1999; Pacyna et al. 2003, EPRI 2000; NESCAUM 2005).

<table>
<thead>
<tr>
<th>Source Category</th>
<th>PHg(%)</th>
<th>RGM(%)</th>
<th>Hg²(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Utility oil boilers</td>
<td>20</td>
<td>30</td>
<td>50</td>
</tr>
<tr>
<td>Municipal waste combustor</td>
<td>20</td>
<td>58</td>
<td>22</td>
</tr>
<tr>
<td>Medical waste incinerator</td>
<td>20</td>
<td>75</td>
<td>5</td>
</tr>
<tr>
<td>Pulp &amp; paper production</td>
<td>20</td>
<td>30</td>
<td>50</td>
</tr>
<tr>
<td>Chlorine production</td>
<td>0</td>
<td>5</td>
<td>95</td>
</tr>
<tr>
<td>Hazardous waste incinerator</td>
<td>22</td>
<td>20</td>
<td>58</td>
</tr>
<tr>
<td>Primary &amp; secondary metal production</td>
<td>10</td>
<td>10</td>
<td>80</td>
</tr>
<tr>
<td>Municipal landfills</td>
<td>10</td>
<td>10</td>
<td>80</td>
</tr>
<tr>
<td>Coal-fired electric utilities – U.S. average</td>
<td>10%</td>
<td>40%</td>
<td>50%</td>
</tr>
<tr>
<td>Coal-fired electric utilities in the Northeast</td>
<td>2%</td>
<td>68%</td>
<td>30%</td>
</tr>
</tbody>
</table>
Table 3. Water chemistry indicators of lakes with elevated concentrations of Hg in fish. **This could be incorporated into the text and the table deleted**

<table>
<thead>
<tr>
<th>Indicator</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>DOC</td>
<td>&gt; 4.0 mg C/L</td>
</tr>
<tr>
<td>pH</td>
<td>&lt; 6.0</td>
</tr>
<tr>
<td>Acid neutralizing capacity</td>
<td>&lt; 100 μeq/L</td>
</tr>
<tr>
<td>Total phosphorus</td>
<td>30 μg/L</td>
</tr>
</tbody>
</table>

Table 4. Mercury exposure to biota in the Northeast – Dave will revise this and present data by functional feeding group

<table>
<thead>
<tr>
<th>Organism</th>
<th>Range</th>
<th>Mean</th>
<th>Effects threshold</th>
<th>% above threshold</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zooplankton</td>
<td>16 to 7,477 ng/g DW</td>
<td>unknown</td>
<td>Adult blood = 3.0 ppm</td>
<td></td>
</tr>
<tr>
<td>Crayfish</td>
<td>23 to 550 ng/g</td>
<td>unknown</td>
<td>Egg = 1.3 ppm</td>
<td></td>
</tr>
<tr>
<td>Common loon</td>
<td></td>
<td></td>
<td>Juvenile blood = 0.7 ppm</td>
<td></td>
</tr>
<tr>
<td>Bald eagle</td>
<td></td>
<td></td>
<td>Adult blood = 3.0 ppm</td>
<td></td>
</tr>
<tr>
<td>Yellow perch &amp; brook trout</td>
<td></td>
<td></td>
<td>Whole body = 0.16 ppm (ecological effects)</td>
<td></td>
</tr>
<tr>
<td>Mink</td>
<td>0 to 69 ppm</td>
<td>20.7 ppm</td>
<td>Fur = 20 ppm</td>
<td>29</td>
</tr>
<tr>
<td>Otter</td>
<td>0 to 234 ppm</td>
<td>20.1 ppm</td>
<td>Fur = 20 ppm</td>
<td>61</td>
</tr>
</tbody>
</table>
Table 5. Results of MCM-HD simulations for Sunday Lake watershed. Shown are results for total mercury in lake water, 3+ year yellow perch and loon blood to the base case and 15% and 50% decreases in total Hg deposition.

<table>
<thead>
<tr>
<th>Media</th>
<th>Base conditions</th>
<th>Simulation period</th>
<th>15% decrease in total Hg deposition</th>
<th>50% decrease in total Hg deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water (ng/L)</td>
<td>0.86</td>
<td>10 years</td>
<td>0.82</td>
<td>0.71</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50 years</td>
<td>0.79</td>
<td>0.63</td>
</tr>
<tr>
<td>Fish (µg/g)</td>
<td>1.14</td>
<td>10 years</td>
<td>1.08</td>
<td>0.94</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50 years</td>
<td>1.04</td>
<td>0.83</td>
</tr>
<tr>
<td>Loon (MLU)</td>
<td>14.48</td>
<td>10 years</td>
<td>14.48</td>
<td>12.67</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50 years</td>
<td>11.25</td>
<td>13.96</td>
</tr>
</tbody>
</table>

Figure 1: Different emission speciation profiles by State (based on ICR data: Required ~85 coal-fired utility units to test for speciated mercury emissions). **Probably not necessary**
Figure 2. Changes in historical deposition of Hg to sediments in West Pond and Little Echo Pond in the Adirondack region of New York from 1820 to the present (after Lorey and Driscoll 1999). The sediment patterns reflect changes in Hg emissions and deposition over time.

Should we think about including the retention time figure that you created, I’ve added a sentence in the text we could link it to.
Figure 3: Yearly Hg wet deposition flux measured at MDN sites in Northeastern U.S.
Figure 4. Average water Hg concentrations within 18-minute grid cells for lakes and streams across northeast North America. Inset figure shows the distribution of Hg concentrations comprising the mean for each quantile.
Figure 5. Box and whisker plots of concentrations of Hg in water and aquatic biota in eastern North America. Also shown are the ranges of percent of total Hg occurring as MeHg. All values were obtained from NERC data and represent wet weight, except for phytoplankton which were obtained from….  
| Somewhat redundant with Table 4 |
Figure 6. Relationship between Hg concentrations in standard length yellow perch and total phosphorus concentration in lakes (A) and box and whisker plots of concentrations of dissolved organic carbon (DOC; B), pH (C), acid neutralizing capacity (ANC; D) for waters in the northeastern U.S. which contain average concentrations of standard length yellow perch that are < 0.3 µg Hg/g.
Figure 7. Mean and standard deviation of Hg concentrations of 13 species of fish in eastern North America (after Kamman et al. 2005). Shown is the EPA criterion for fish Hg concentrations.

NOTE: 0.3 ppm is not denoted; do we need three significant figures on the x-axis?